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INVESTIGATION OF THE SURFACE STATE OF NICKEL AND IRON CATHODES  
FROM THE DROP IN POTENTIAL AFTER DISCONNECTING THE CURRENT

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INVESTIGATION OF THE SURFACE STATE OF NICKEL AND IRON CATHODES  
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ABSTRACT: Oscillographic curves for the drop in potential after disconnecting the current are used for studying the surface state of cathodically polarized nickel and iron. It is shown that the capacitance of nickel and iron cathodes, as computed from the curves for the potential drop, depends on the electrode potential and on the time elapsed from the instant when the polarizing current is switched off.

It may be assumed that the minimum capacitances of 19-24  $\mu\text{f}/\text{cm}^2$  for a nickel cathode in acid and alkali solutions and 27-28  $\mu\text{f}/\text{cm}^2$  for an iron cathode in acid solutions are close to the capacitances for the double layer on these metals. The capacitance of an iron electrode in alkali solutions and for high cathodic polarizations is appreciably greater than the capacitance of the double layer.

The increase in capacitance as the cathode potential drops is due to the reaction of ionization of adsorbed electrochemically active hydrogen, the quantity of which increases with a reduction in cathodic polarization. The relatively low capacitances as compared with that of a hydrogen condenser plate show that only a small area of the nickel and iron electrode surface is covered with adsorbed hydrogen which can be ionized in the given potential range. Surface coverage by electrochemically active hydrogen is low in an acid medium and somewhat greater in an alkali solution, although it still does not exceed 0.03 for a nickel cathode, and 0.20 for iron.

The investigation of the surface state of nickel and iron by measuring the differential capacitance of an electrode with the use of variable current or from curves for the potential drop after switching off the polarizing current [1-3] have been limited by a comparatively low cathode potential. Because of experimental difficulties, the excess voltage region above 0.3 v was not studied in the reference works listed above. Thanks to an improvement in equipment and a reduction in the time interval over which the electrode potential is measured, we have been successful in appreciably raising the cathode potential at which the electrode capacitance can be determined.

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\* Numbers in the margin indicate pagination in the foreign text.

## Experimental Part

The equipment and measurement method were outlined in a previous work [4].

The nickel electrode was a plate of spectrally pure nickel with a surface area of  $0.2 - 0.5 \text{ cm}^2$ . After thorough scouring with finely powdered glass, degreasing in hot concentrated alkali and washing with water, the electrode was fired in a stream of hydrogen at  $420-450^\circ\text{C}$  in the measurement cell itself. Firing lasted for 3-5 hours. After the cell had been cooled, the electrode was immersed in the solution without coming into contact with the air.

An iron electrode of spectrally pure iron, cylindrical in shape with a diameter of 1.15 mm and a working surface area of  $0.2 - 0.4 \text{ cm}^2$  was freed from surface oxides [5] by firing in a hydrogen stream at  $700^\circ\text{C}$  for several hours followed by evacuation at  $900^\circ\text{C}$  for 1 hour. The electrode thus prepared was kept in a test tube with a ground-joint stopper. The electrode was not fired immediately preceding the experiment.

The alkali solutions were prepared either by dissolving prewashed sticks of chemically pure alkali NaOH or KOH in doubly distilled water, or by using the same medium for decomposing an electrochemically produced potassium amalgam. Experiments showed that solutions produced from chemically pure NaOH and KOH or from the amalgam, after protracted purification on a cathode of the same material as that used for the electrode to be studied, yielded comparable results. Preliminary purification of the solution was done for ten hours at a current density of  $10^{-2} \text{ a/cm}^2$ .

The electrode potential was measured with respect to a hydrogen electrode in the same solution. Measurement was done after cathodic polarization of the specimen electrode for one hour at a current of about  $10^{-2} \text{ a/cm}^2$ . The electrode potential was rapidly established in acid solutions, and remained stable throughout the entire experiment. In the case of alkali solutions, there was a slow shift in the potential of nickel and iron electrodes toward the negative side, which sometimes continued even after ten hours of electrode polarization at constant current density. The experiments which we conducted showed that the contaminants present in the alkali solution have a strong effect on the increase in overvoltage during protracted polarization. The phenomena associated with an increase in overvoltage during protracted polarization are not considered in this article.

The results of individual measurements of the excess voltage on nickel and iron under identical experimental conditions were repeatable within limits of 10-20 mv. To prevent dissolution of the electrode in acid or oxidation in alkali, the region of low cathode potentials was not investigated. After plotting a repeatable excess voltage curve, oscillographs of the potential drop were taken at various polarizing current densities. The

electrode capacitance was calculated from equation (3) given in [4].

*Determining the Capacitance of a Nickel Cathode in Alkali Solutions.* Data on determination of nickel capacitance in acid solutions were given in [6].

/1231

In alkali solutions (0.6 - 0.8 *N* NaOH, 1.2 *N* KOH and 5 *N* KOH), it was found that the hydrogen overvoltage on nickel was a linear function of the logarithm of polarizing current density at low and moderate densities (up to  $10^{-2}$  a/cm<sup>2</sup>) with a proportionality factor equal to 0.10 - 0.11 v. At current densities higher than  $10^{-2}$  a/cm<sup>2</sup>, it was found that the factor *b* increases with current density. In computing electrode capacitance, the value of this factor was determined from experimental overvoltage curves (with regard to correction for the ohmic potential drop in the solution).

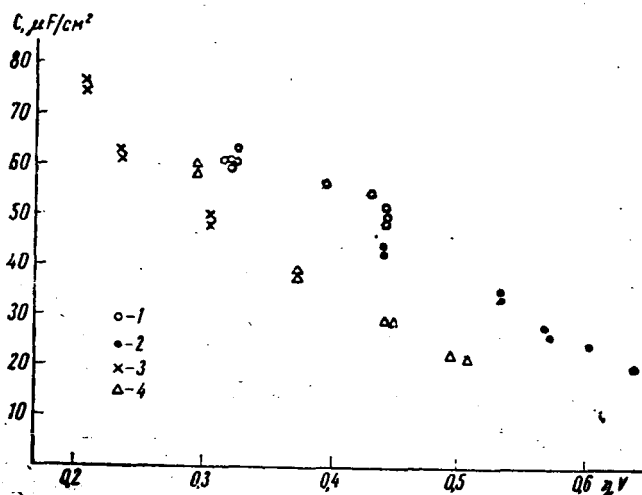


Figure 1. Initial Capacitance of Nickel Electrode as a Function of Excess Voltage. 1--0.62 *N* NaOH; 2--0.65 *N* NaOH; 3--0.8 *N* NaOH; 4--5 *N* KOH.

Shown in Figure 1 is the capacitance of a nickel electrode calculated from the initial section of the potential drop curve as a function of the excess voltage in 0.6-0.8 *N* NaOH and 5 *N* KOH. This figure shows that the initial capacitance of the electrode depends on potential: when  $\eta = 0.6$  v,  $C = 23 \mu\text{f}/\text{cm}^2$  of the apparent surface; with a reduction in excess voltage to 0.2 v,  $C$  increases to  $75 \mu\text{f}/\text{cm}^2$ .

The results of computation of nickel capacitance for individual moments of time after disconnecting the polarizing current show that the increase in capacitance with time *t* corresponds in the first approximation to the increase in capacitance computed from the initial sections of oscillographs for the potential drop with a reduction in excess voltage.

Surface preparation of the nickel electrode affects its behavior in alkali solutions. Repeatable capacitances were obtained only after identical pretreatment of the surface of the specimen. Experiments conducted with an electrode previously cathodically polarized in 0.1 *N* H<sub>2</sub>SO<sub>4</sub> without firing in a hydrogen atmosphere showed considerably higher capacitances ( $C = 70 \mu\text{f}/\text{cm}^2$

in 0.6 N NaOH at  $\eta = 0.5$  v, while  $C = 130 \mu\text{f}/\text{cm}^2$  when  $\eta = 0.2$  v), although the nature of the dependence of capacitance on potential is identical for both electrodes.

*Determining the Capacitance of an Iron Cathode in Acid and Alkali Solutions.* The  $\eta$ -log  $i$  relationship for iron in 1 N KOH is linear in the region of current densities from  $10^{-2}$  to  $10^{-1}$  a/cm<sup>2</sup>, the constants in the Tafel equation being  $a = 0.70$  v and  $b = 0.12$  v.

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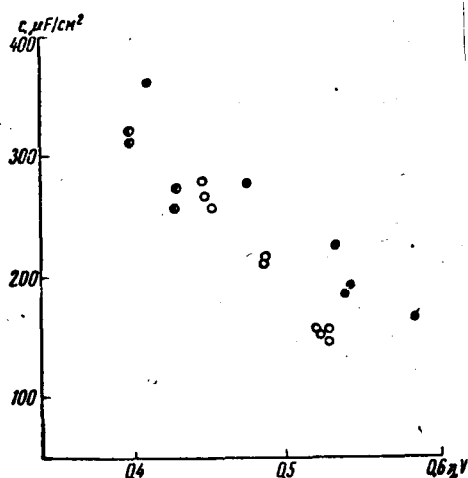


Figure 2. Initial Capacitance of Iron Electrode As a Function of Excess Voltage in 1 N KOH. The Different Symbols Refer to Different Experiments.

Shown in Fig. 2 are the results of three experiments in determining the initial capacitance of an iron cathode at various potentials in a 1 N solution of KOH. Even at high excess voltages, the capacitance of the electrode is 150-200  $\mu\text{f}/\text{cm}^2$ . The capacitance of an iron electrode increases with time  $t$ , this increase is more appreciable than the increase in the initial capacitance with a reduction in cathode potential.

The potential drop for an iron cathode in a 1 N solution of  $\text{H}_2\text{SO}_4$  was measured only in the narrow range of current densities from  $5.4 \cdot 10^{-3}$  to  $7.4 \cdot 10^{-2}$  a/cm<sup>2</sup>. The results of determination of capacitance from the initial segments of curves for the potential drop at various polarizing current densities show that the capacitance of the electrode

increases with a reduction in excess voltage from 27-28  $\mu\text{f}/\text{cm}^2$  at  $\eta = 0.5$  v to 60  $\mu\text{f}/\text{cm}^2$  at  $\eta = 0.38$  v (Fig. 3). The capacitance of an iron cathode in an acid solution increases with time  $t$ .

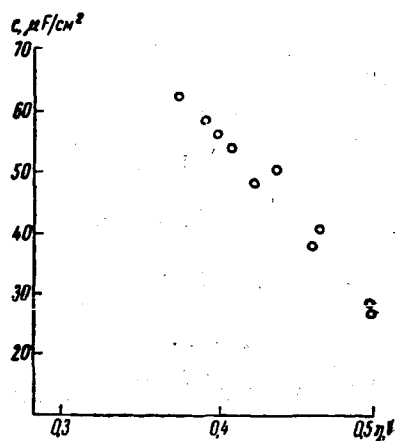


Figure 3. Initial Capacitance of an Iron Electrode as a Function of Excess Voltage in 1 N  $\text{H}_2\text{SO}_4$ .

## Discussion of the Results

The results of measurement of the potential drop curves for nickel and iron cathodes in acid and alkali solutions show a difference in their behavior as compared with that of mercury [4]: it was found that the electrode capacitance calculated from the experimental curve is dependent on the potential and on the time elapsed from the instant when the current is disconnected. This dependence cannot be explained by assuming that the excess voltage is due to the slowness of the discharge reaction on the surface, which is practically free from adsorbed hydrogen. It may be expected that the slowness of reactions of eliminating atomic hydrogen plays a greater part in metals with a higher hydrogen adsorption energy, specifically nickel and iron, than in the case of mercury.

Let us examine in more detail the case where excess voltage is caused by the slowness of eliminating adsorbed hydrogen and the discharge reaction rate is high, so that the equilibrium between adsorbed hydrogen and adsorbed solution is not upset when the current passes through. Given below is A. N. Frumkin's derivation of the equation corresponding to the curve for the potential drop after the current is switched off, according to recombination theory.

We shall assume that the electrode potential  $\phi$  is dependent on the concentration of hydrogen atoms in accordance with the expression /1233

$$\phi = \text{const} - \frac{RT}{F} \ln [H]. \quad (1)$$

If the recombination rate for atomic hydrogen is proportional to the square of its concentration  $[H]$ , then the rate of change in concentration after switching off the current is determined by the equation

$$\frac{d[H]}{dt} = -q[H]^2, \quad (2)$$

where  $q$  is the rate constant for the reaction of recombination.

After integrating equation (2), we get the expression

$$\frac{1}{[H]} = qt + \frac{1}{[H_0]}. \quad (3)$$

where  $[H_0]$  is the value of  $[H]$  when  $t = 0$ .

It follows from equations (1) and (3) that

$$\varphi = \text{const} + \frac{RT}{F} \ln \left( qt + \frac{1}{[H_0]} \right). \quad (4)$$

Hence the change in electrode potential after disconnecting the current is equal to

$$\varphi_t - \varphi_0 = \frac{RT}{F} \ln (1 + q [H_0] t), \quad (5)$$

where  $\varphi_0$  is the value of  $\varphi$  when  $t = 0$ .

The density of the polarizing current is  $i_0 = qF[H_0]^2$ . Substituting in equation (5), we get

$$\varphi_t - \varphi_0 = \frac{RT}{F} \ln \left( 1 + \frac{i_0 t}{F[H_0]} \right). \quad (6)$$

Let us call the quantity  $C_s = -F \frac{\partial [H]}{\partial \varphi}$  the capacitance of the electrode with respect to hydrogen. It follows from equation (4) that

$$\frac{\partial [H]}{\partial \varphi} = -\text{const} \frac{F}{RT} e^{-F\varphi/RT} = -\frac{F}{RT} [H]. \quad (7)$$

According to the definition of capacitance and expression (7),

$$C = \frac{F^2}{RT} [H]. \quad (8)$$

At the instant of current interruption  $t = 0$ ,  $C = C_0$

$$C_0 = \frac{F^2}{RT} [H_0]. \quad (9)$$

From equations (6) and (9), it follows that

$$\varphi_t - \varphi_0 = \frac{RT}{F} \ln \left( 1 + \frac{i_0 t}{\frac{RT}{F} C_0} \right) = 2b \ln \left( 1 + \frac{i_0 t}{2b C_0} \right), \quad (10)$$

where  $b = RT/2F$  as a consequence of recombination theory.

Formula (10) is derived on the basis of the simplest form of recombination theory, which may be applied only when the surface coverage for the electrode with respect to adsorbed hydrogen atoms is extremely low. Cathodic liberation of hydrogen in metals where the energy of the Me-H bond is high can take place only on the surface of an electrode which is covered to a considerable extent by adsorbed atoms. In this case, the kinetic and thermodynamic activities of the adsorbed hydrogen cannot be considered proportional to its surface concentration as was done above (equations (1) and (2)).

A. N. Frumkin and A. I. Shlygin [7] showed that in the case of moderate surface coverages, the potential of a platinum electrode is an approximately linear function of the quantity of adsorbed hydrogen, which corresponds with a logarithmic relationship between the adsorbed quantity and the pressure of the hydrogen. The logarithmic isotherm may be theoretically derived if certain assumptions are made on the state of the adsorbed hydrogen on the electrode [8]. In this case, a formula which may be used is that derived in the work by A. N. Frumkin and N. A. Aladzhhalova for the section of the experimental curve which was caused by slow elimination of molecular hydrogen (equation (13) in reference work [9]): /1234

$$\varphi_t - \varphi_0 = \frac{RT}{2F\beta'} \ln \left( 1 + \frac{2\beta' i_0 t}{NF} \right), \quad (11)$$

where  $\beta$  and  $f$  are constants and  $N$  is the factor by which the coverage must be multiplied to obtain the surface concentration of hydrogen in gram atoms per square centimeter.

In this case, the electrode capacitance with respect to adsorbed hydrogen is expressed by the formula

$$C = \frac{NF^2}{RTf} \quad \text{or} \quad C = \frac{F^2}{RTf\theta} [H], \quad (12)$$

where  $\theta$  is surface coverage by hydrogen atoms.

According to formulas (11) and (12), the curve for potential drop is expressed by the equation

$$\varphi_t - \varphi_0 = \frac{RT}{2F\beta'} \ln \left( 1 + \frac{i_0 t}{\frac{RT}{2F\beta'} C} \right) = b \ln \left( 1 + \frac{i_0 t}{bC} \right), \quad (13)$$

where  $b = RT/2F\beta'$ , as was shown by M. I. Temkin.



Consequently, if the adsorbed hydrogen is in equilibrium with the hydrogen ions (or water molecules) in the solution, and if this hydrogen behaves identically to what we know from the charging curves for a platinum electrode, then the potential drop is defined by equation (13), and the capacitance calculated from the curve for this drop is nearly constant and independent of  $\phi$  or  $t$ .  $C$  is constant since  $\phi$  is a linear function of  $\theta$  in the given case.

It follows from what has been said above that the nature of the potential drop curve cannot yet be used for choosing between the mechanisms of retarded discharge or delayed recombination. Both theories yield identical equations for the potential drop curve, with a difference only in capacitance values: in the first case the capacitance calculated from the drop curve is equal to that of the electric double layer, while in the second case the capacitance is that of a hydrogen condenser plate, which exceeds the capacitance of the electric double layer by a factor of approximately 100. The experimentally observed capacitance of nickel and iron electrodes as related to  $\phi$  and  $t$  contradicts both the mechanism of delayed discharge and that of retarded recombination [10]. Therefore it may be assumed in the case of nickel and iron electrodes that no mechanism in pure form is realized, but rather the process must be considered in stages, all taking place at comparable rates.

The behavior of the curves for potential drop with all possible combinations of assumptions on the rate of the various stages has not yet been successfully calculated. Grahame [11] also points out the complexity of this computation. Some special cases of calculating the behavior of drop curves may be used for explaining the results found in this work.

It was shown in the work by A. N. Frumkin and N. A. Aladzhhalova [9] that when certain assumptions are made concerning the properties of the adsorbed hydrogen in the case of combined delayed discharge and delayed recombination, the excess voltage is the sum of two parts:  $\eta = \eta_1 + \eta_2$ , where  $\eta_1$  is due only to the slowness of the discharge, and  $\eta_2$  is associated only with the slowness of elimination of molecular hydrogen. In this case, there should be two segments on the drop curve, the first showing a capacitance corresponding to that of the double layer, and the second corresponding to that of an atomic hydrogen plate.

A more rapid increase may be expected in  $\eta_1$  as compared with  $\eta_2$  as the excess voltage increases, because of the larger coefficient associated with  $\log i$  in the equations of the delayed discharge theory. If an electrochemical mechanism is responsible for desorption of atomic hydrogen, as is extremely likely at high current densities, the increase in surface concentration of adsorbed hydrogen may stop as a result, and consequently there may also be no more rise in  $\eta_2$  as polarization increases [12]. Therefore, the part played by  $\eta_1$  in the potential drop should increase as the current density /1235 rises, with a resultant increase in the length of the section of the drop curve corresponding to double-layer capacitance.

We have assumed up to this point that the behavior of the adsorbed hydrogen may be completely interpreted on the basis of hypotheses developed from the example of a platinum hydrogen electrode. But in the case of a nickel electrode (and possibly also an iron electrode), the behavior of atomic hydrogen is more complicated. There are areas on the nickel surface with a heat of hydrogen adsorption which differs sharply from that of the remainder of the surface [13, 14]. The discharge and desorption reactions may be retarded to different degrees on different sections of the nickel electrode surface. We may write  $\eta = \eta_1 + \eta_2$  for each individual section, but the ratio of  $\eta_1$  to  $\eta_2$  varies from section to section. Therefore, the overall excess voltage can no longer be expressed as the simple sum  $\eta_1 + \eta_2$ . This means that the sections of the drop curve corresponding to  $\eta_1$  and  $\eta_2$  overlap each other in this case. By a certain time  $t$ , the double layer on some sections of the surface has had time to reach a state of equilibrium with the adsorbed hydrogen, while in other sections this equilibrium has not been achieved. In these cases, the capacitance of the electrode at time  $t$  is not equal to that of the double layer or that of the hydrogen plate, but has some intermediate value.

Let us use the idea of the "electrochemical activity" of the adsorbed hydrogen, considering hydrogen electrochemically active when it is capable at a given potential of ionization at a rate comparable with that of the other reactions involving the adsorbed hydrogen. The potential drop is retarded by ionization of the adsorbed hydrogen with a resultant increase in the capacitance calculated from the drop curve. The discrepancy between the capacitance at a given potential and that of the double layer characterizes the electrochemical activity of the adsorbed hydrogen. It may be assumed that the activity of the adsorbed hydrogen differs on different sections of the cathode surface. The oscillographic curves for the drop may be used in calculating the coverage of the surface by electrochemically active hydrogen, but give no basis for judging the total quantity of adsorbed hydrogen on the metal surface.

The experiment shows that the capacitance of a nickel electrode calculated from oscillographic drop curves falls as the cathodic polarization rises, and approaches  $20 \mu\text{f}/\text{cm}^2$  at extremely high polarizations. This relationship between capacitance and potential is due to a reduction in the amount of electrochemically active hydrogen on the surface as the excess voltage increases. It may be assumed that the minimum values of 19-24  $\mu\text{f}/\text{cm}^2$  of apparent surface for a nickel electrode are close to the capacitance of the electric double layer. The surface area over which the adsorbed hydrogen maintains equilibrium with the solution at these potentials cannot be greater than a few thousandths of the total surface area. If we take the capacitance of  $17.7 \mu\text{f}/\text{cm}^2$  for the perfectly smooth negatively charged surface of mercury as a reference point [4], then the capacitances which we have found for a nickel electrode seem even somewhat underestimated. A possible explanation for this phenomenon was given previously [6].

For identical excess voltage, the capacitance of a nickel electrode in

an alkali solution is considerably greater than that in an acid solution. When  $\eta = 0.45$  v, the capacitance in 1 N  $H_2SO_4$  is  $22 \mu f/cm^2$ , corresponding to a surface coverage by electrochemically active hydrogen of  $\theta \sim 10^{-3}$  or less\*, while in 0.6 - 0.8 N NaOH at  $\eta = 0.45$  v, the capacitance is  $40-50 \mu f/cm^2$  and  $\theta \sim 10^{-2}$ . As the excess voltage decreases,  $\theta$  increases; at  $\eta = 0.2$  v, the surface coverage is equal to  $10^{-2}$  for an acid solution and  $3 \cdot 10^{-2}$  for an alkali solution. The difference in the quantity of adsorbed hydrogen on a nickel electrode in alkali and acid solutions is probably due to an increase in the strength of the Ni-H bond with a transition from acid solutions to alkali. Similar behavior of the strength of the bond between a metal atom and an adsorbed hydrogen atom as a function of the composition of the solution was previously observed in the case of a platinum electrode [7].

Protracted firing of the nickel electrode at a temperature of  $450^\circ C$  apparently changes the surface state of the specimen. Due to recrystallization, there is a change in the distribution of the surface into sections with varying heats of adsorption of atomic hydrogen, resulting in an experimentally observed change in the electrode capacitance. This hypothesis agrees with the results of adsorption measurements [15].

Higher capacitances were found for an iron electrode in an alkali solution. This cannot be attributed to the greater surface roughness of iron. Measurements of the drop in excess voltage of an iron electrode in an acid solution showed that the capacitance at high excess voltages (0.5 v) is  $27-28 \mu f/cm^2$  of the apparent surface. This is apparently close to the capacitance of the double layer on iron. The amount of electrochemically active hydrogen on the surface of iron in alkali solutions is considerably greater than in acid solutions: at an excess voltage of 0.55 - 0.6 v in a 1 N solution of KOH,  $\theta$  amounts to 0.05 - 0.1, while at  $\eta = 0.4$  v,  $\theta \sim 0.15 - 0.2$ . A difference in the behavior of adsorbed hydrogen on iron in acid and alkali solutions in the case of cathodic polarization was observed by I. A. Bogatskaya [16] from measuring the excess voltage sent through a thin iron plate.

The results of this work show that excess voltage for the greater part of the surface of iron and nickel electrodes cannot be interpreted as the equilibrium potential of adsorbed hydrogen. The portion of the surface on which adsorbed hydrogen is in equilibrium with hydrogen ions (or water molecules) is insignificant in comparison with the entire surface. Only on an iron electrode in an alkali solution is the fraction of these sections greater, but even in this case it is no more than 10-20% of the entire surface area. Even though there is adsorbed hydrogen on the remainder of the surface, it is not electrochemically active at appreciable overvoltages. On

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\* It is assumed in calculating the coverage  $\theta$  that the capacitance of the electric double layer is  $20 \mu f/cm^2$ , while that of a hydrogen plate is  $2,000 \mu f/cm^2$ .

these sections, which comprise the greater part of the surface, the discharge stage for a hydrogen ion or water molecule is characteristically irreversible in the process of hydrogen liberation.

However, we cannot draw an unambiguous conclusion on this basis concerning the mechanism of hydrogen liberation on nickel and iron electrodes as a whole. We do not know the distribution of current between sections with adsorbed hydrogen of higher or lower electrochemical activity. A unique explanation of the mechanism responsible for hydrogen liberation on metals of the iron group demands comprehensive utilization of research data obtained by using various experimental methods.

We are sincerely grateful to Academician A. N. Frumkin for his constructive criticism during the work and for his consideration of the results.

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